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Use of Miniature Glass-Needle-Type TL Dosimeters in Finger-Ring Applications

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ABSTRACT

Miniature glass-encapsulated needle-shaped thermoluminescent (TL) dosimeters, 1.4 mm in outer diameter and 12 mm in length, have been investigated for routine application in finger rings to monitor the dose received by the hands, e.g., in certain Linac repair operations. Both $\text{CaF}_2:\text{Mn}$ and LiF-filled dosimeters were studied, and special attention was given to the simplification or elimination of the cumbersome annealing procedures normally used with LiF dosimeters.

It was found that the heights of the LiF TL glow peaks in the vicinity of 200°C , usually employed for dosimetry, display a marked growth as a function of storage time occurring either before or after a given γ -ray exposure. This is probably caused by the migration and accumulation of simple, low-temperature trapping centers into more complex, higher-temperature centers, as suggested by some other investigators. The magnitude of the effect depends on the population of low-temperature traps present, and on the readout heating rate employed. It was found possible to reduce the dosimetry error caused by this sensitivity increase to negligible levels ($\pm 3\%$) through the use of a routine, pre-readout annealing treatment of 15 min at 100°C . No additional annealing procedure was found to be necessary for LiF, so long as the γ -ray exposures were not large enough ($\sim 10^3$ R) to cause radiation-induced sensitization of the phosphor. $\text{CaF}_2:\text{Mn}$ showed no significant change in sensitivity either upon storage or radiation exposure.

PROBLEM STATUS

This is an interim report on one aspect of the problem; work is continuing on dosimeter evaluations and problems.

AUTHORIZATION

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USE OF MINIATURE GLASS-NEEDLE-TYPE TL DOSIMETERS IN FINGER-RING APPLICATIONS

INTRODUCTION

The miniature glass-encapsulated "needle"-type thermoluminescent dosimeter (TLD), originated by Ginther and Attix (1) at the Naval Research Laboratory (NRL) in 1960, has found increasing application, primarily for medical dosimetry and other applications where exposures generally exceed approximately 1 R, the practical lower limit of one such commercially available dosimeter. This dosimeter contains 0.6 mg of phosphor, powdered $\text{CaF}_2:\text{Mn}$ or LiF (TLD-100 or TLD-700), sealed in an evacuated glass capillary tube 0.8 mm in outside diameter and 6.0 mm long. In the exposure range from 1 to 1000 R it was found to have a precision of ± 0.1 R or $\pm 3\%$, whichever is greater (2). Because of its inability to measure exposures less than 1 R with sufficient precision, it has little usefulness as a personnel monitoring dosimeter. However, other larger prototype models up to 1.5 mm in diameter and 15 mm long also were made at NRL (3), and these are capable of detecting exposures of 10 mR. Their present commercial availability makes feasible their routine use in such applications as finger-ring extremity dosimetry.

The present investigation began in 1967. By that time the NRL Linac had been in operation long enough so that routine maintenance necessitated working on radioactive accelerator parts, causing significant radiation exposures to the hands of personnel. Since previous experience at NRL had shown that the use of film in finger rings gave unreliable results, primarily due to light leakage, it was decided to use a TLD system. The absence of significant beta exposures (due to the usual time lapse* between accelerator shut-down and start of maintenance) and a desire for more accurate and easily handled dosimeters, led us to consider using the TL needle dosimeters rather than the more commonly used system based on Teflon impregnated with phosphor (4). Desiring a simple, routine procedure for personnel monitoring, we decided to investigate the use of LiF and $\text{CaF}_2:\text{Mn}$ -filled dosimeters without any pre- or postexposure annealing procedures. The cumbersome annealing process normally recommended for LiF is one of the major deterrents to its use in health physics applications. Hence, simplification or elimination of these procedures was a major objective, and comprised a large part of this effort. It was thought that employing the glow-peak height rather than the light sum might be advantageous in this connection. Some of the results obtained were quite unexpected and interesting, perhaps shedding some additional light on the TL mechanisms in LiF .

THE DOSIMETRY SYSTEM

Initial investigations of the LiF and $\text{CaF}_2:\text{Mn}$ needle dosimeters were made using laboratory-made dosimeters containing 15 to 20 mg of phosphor in glass needles 1.5 mm in diameter and 15 mm long. When needles 1.4 mm in diameter and 12 mm in length

*Time lapse from shut-down to maintenance varies depending on running time and the particular mode of operation. It is usually not less than 4 hours. Routine maintenance is performed on Mondays after the machine has been shut down for the weekend.

became commercially available, these were adopted and all the results given here were obtained using these dosimeters. Included in the testing were three different batches of ^7LiF (TLD-700) needles, three of ^6LiF (TLD-600) and two of $\text{CaF}_2:\text{Mn}$. Typically, the LiF needles contained 9 to 10 mg of phosphor, while the $\text{CaF}_2:\text{Mn}$ needles contained 14 to 15 mg.

The dosimeters were read on an EG&G Model 2 TLD reader,* using a locally made heating element consisting of eight turns of No. 24 Chromel wire soldered to the base of an EG&G Model TL-32 glass bulb dosimeter from which the glass envelope and detector element had been removed (see Fig. 1). To insure good thermal coupling between the heating coil and the glass needles the coil was formed by winding the wire on the shank of a No. 55 drill (1.321 mm diam), then drilling the inside of the coil with a No. 53 drill (1.511 mm diam), thus forming a D-section wire with the flat side toward the dosimeter. This heating element was plugged into the standard EG&G reading head before being inserted into the reader. Both the LiF and $\text{CaF}_2:\text{Mn}$ dosimeters were normally read using a constant heater current of 4.0 A. The higher-temperature glow peak of the latter necessitated extending the usual 13-sec heating period of the Model 2 reader by an additional 4 sec. Efforts to reduce the readout time of the $\text{CaF}_2:\text{Mn}$ needles by increasing the heater current resulted in excessive "heat signal" from incandescence of the heating coil, which raised the minimum detectable exposure of the dosimeters. For some of the laboratory tests a Honeywell Electronik 19 recorder was substituted for the less-sensitive built-in recorder of the Model 2 reader to extend the range of the instrument one decade and thus to achieve greater precision in reading small exposures.

For the personnel-monitoring aspects of the tests the commercially available plastic finger ring shown in Fig. 1 was used. This ring normally uses film as the radiation detection element and is supplied with cadmium filters for energy compensation. We replaced the cadmium filters with a disc of aluminum 13.50 mm in diameter and 2.38 mm thick, into the edge of which were drilled two No. 53 holes 12 mm deep to accept the needle TLD's.

RESULTS OF LABORATORY TESTING AND EVALUATION

Unless otherwise specified, all the following results were obtained with no pre- or postexposure annealing other than cycling the dosimeter through the Model 2 reader, either to read a previously exposed dosimeter or to simulate such a reading procedure.

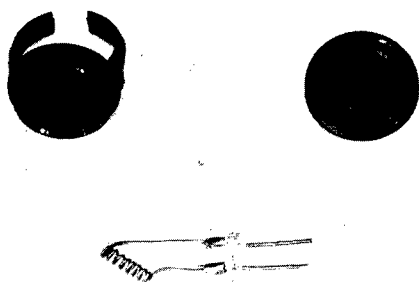


Fig. 1 - Above: Disassembled commercially available finger ring used for the field tests, showing needle dosimeters in the aluminum insert disk. Below: Coiled wire heater used to read the needles in the commercial TLD reader.

*Nitrogen flow is not employed in the sample chamber of this reader.

Dosimeters were immediately removed from the reading head following readout; hence, they rapidly cooled to room temperature in about 20 sec. Unless otherwise specified, the major glow-peak height was used as the measure of the exposure.

General Characteristics

The glow curve shown in Fig. 2 was obtained using a dosimeter from the first batch of ^7LiF dosimeters purchased, which we will call batch a. The dosimeter was given a ^{60}Co γ -ray exposure of 1 R in 1 min and read less than 1 min afterward. The glow peaks have been numbered in ascending temperature to correspond with others' notation (5). Peak 1 was rarely observed, since it fades rapidly at room temperature, and was already negligible by the time our usual "prompt" readings were made, ~ 10 min after exposure. For the later batches of LiF dosimeters, the heights of peaks 2, 3 and 4 were relatively greater, with peaks 3 and 4 in some cases exceeding peak 5. The height of peak 6 (probably partially "spurious" signal due to tribothermoluminescence) varied from batch to batch and run to run, usually being lower than that shown here. The average sensitivity, e.g., peak-height response per roentgen, of the LiF dosimeter batches varied considerably (as much as 50%), with the ^6LiF dosimeters generally, but not always, being the less sensitive. This, along with the differences in glow-curve shape, necessitated keeping the different batches separate.

The glow curve for the $\text{CaF}_2:\text{Mn}$ dosimeters was a broad single peak with the maximum occurring 14 sec after the beginning of the heating cycle. The two dosimeter batches obtained showed the same radiation sensitivity per roentgen, which was about four times that of the LiF dosimeters when read on the Model 2 reader. Because of interference from the "heat signal," caused by the higher temperatures reached in the longer heating cycle required to read the $\text{CaF}_2:\text{Mn}$ dosimeters, the practical minimum detectable exposure was about 10 mR, the same as could be achieved with the LiF dosimeters.

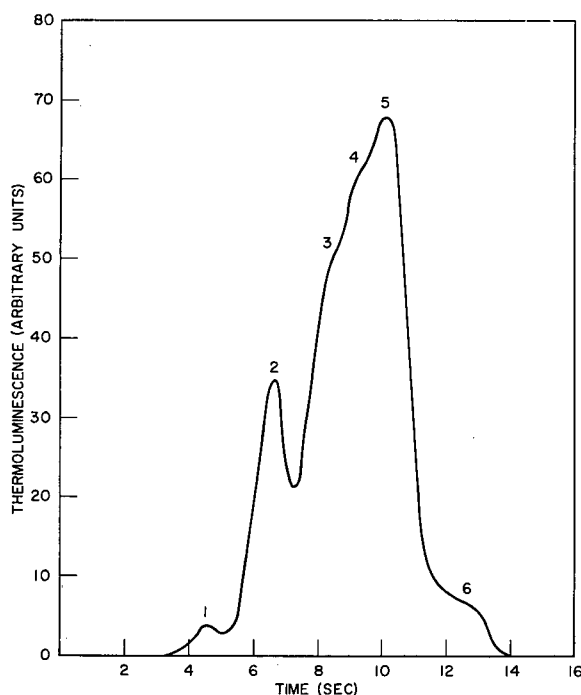


Fig. 2 - Glow curve (thermoluminescence intensity vs heating time) for a ^7LiF needle dosimeter of batch a given a 1 R ^{60}Co γ -ray exposure in 1 min and read < 1 min later. Dosimeter was annealed just prior to exposure by cycling it through the TL reader.

Change of Glow Curve on Storage Before Irradiation

Figure 3 illustrates one important effect of not using any annealing procedures, other than the normal readout heating cycle itself, with the LiF needles. If, following a normal readout operation, various periods of time are allowed to elapse in dark storage at room temperature before irradiating and promptly reading out the dosimeters, one observes a progressive decrease in the low-temperature glow peaks and an increase in the high-temperature peak-height maximum, with a net reduction in total glow-curve area. The more pronounced the low-temperature peak heights are in a given batch of needles, the more marked is this time dependence of the glow-curve shape. Each of the six batches of LiF dosimeters tested showed a different low-temperature glow-curve structure. The glow curve shown in Fig. 2 was obtained using a dosimeter from the batch a evidently having the smallest concentration of low-temperature traps, whereas Fig. 3 was obtained with a dosimeter taken from a batch b having the greatest concentration of low-temperature traps. Note that with prompt exposure and readout the height of peak 2 varies among the batches from about 5/10 to 8/10 the height of peak 5. By using the University of Wisconsin "standard" preannealing procedure (1 hr at 400°C plus 24 hr at 80°C) the low-temperature structure of any of the LiF dosimeters could be reduced to a very low level.

The relatively pronounced low-temperature glow peaks observed when irradiation occurs promptly after a prior readout are evidently caused by the rapid cooling of the dosimeters after removal from the reader. We were able to get a similar, but slightly smaller effect in dosimeters rapidly cooled from 400°C by dropping them on a piece of aluminum foil. The effect of cooling rate on glow curve shape has been previously investigated (5), and it has been suggested (6) that the rapid cooling may be "freezing in" the equilibrium distribution of the impurities and associated trapping sites that exist at

the higher temperatures, whereas slow cooling allows the formation of aggregates of the simpler impurity centers. Room-temperature storage for long periods tends to simulate the effect of slow cooling. The curve (C) shown for 1 month in Fig. 3 is in fact similar in shape to that obtained using the standard preexposure annealing procedure, except that the major peak height in that case is 5 to 10% lower.

The ratios of the peak height maxima of the stored batch b dosimeters to the same ones subsequently dosed and read promptly after readout cycling were as follows for the various storage times: 24 hr, 1.04; 4 days, 1.14; 1 week, 1.23; 2 weeks, 1.31; and 1 month, 1.38. These data are plotted in Curve A, Fig. 4.

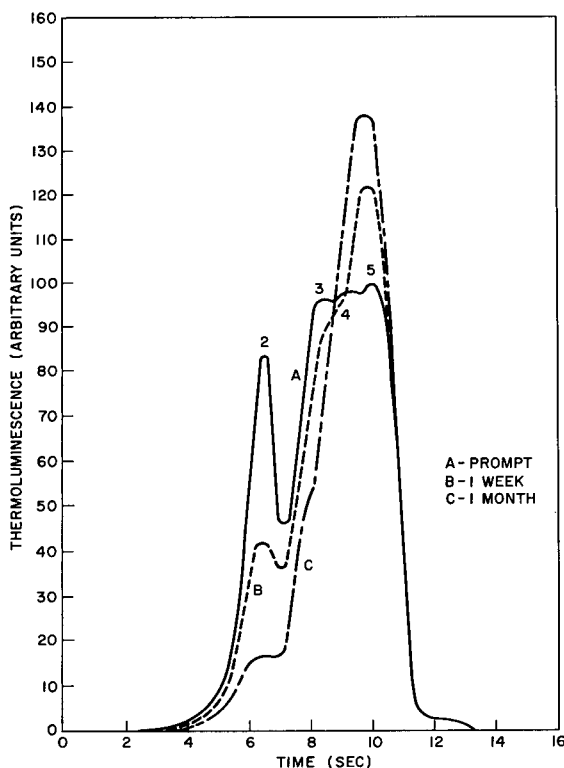


Fig. 3 - Glow curves (thermoluminescence intensity vs heating time) for a ${}^7\text{LiF}$ needle dosimeter of batch b stored in darkness at 22°C for various times since the last readout, then exposed to 12.5 R of ${}^{60}\text{Co}$ γ radiation at 2.5 R/min and read promptly (10 min) after exposure.

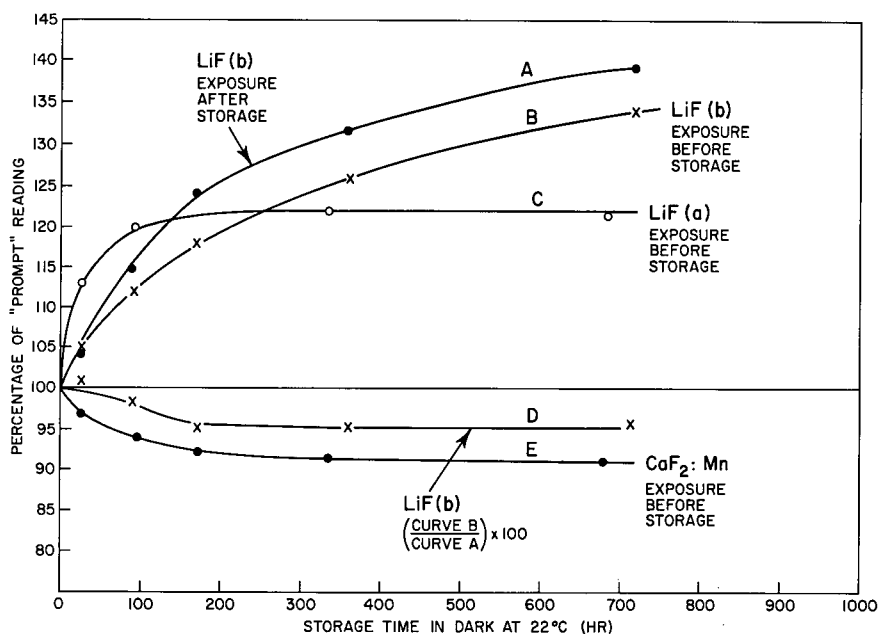


Fig. 4 - Variation of maximum glow-peak heights as a function of storage time in darkness at 22°C. Points represent averages of 10 dosimeters. Immediately after the TL readout cycle, the dosimeters were again given the same exposure (12.5 R of ⁶⁰Co γ rays delivered at 2.5 R/min) and read out 10 min later. The (stored/prompt) readings × 100 are the quantities plotted, except for Curve D which is the ratio of (Curve B/Curve A) × 100.

LiF dosimeters having smaller low-temperature peaks showed a lesser increase in peak height, the least ratio for a month of storage being 1.23 for batch a, stored/prompt. Changes in glow-curve shape, similar to those obtained by storage at room temperature, were also obtained by annealing the dosimeters for various times at 80°C or 100°C before exposure. Annealing at 100°C for 15 min produced about the same change as 1-month storage at room temperature, as will be shown later.

No corresponding change in glow-curve shape or peak height with preexposure storage time after the last readout was observed with the CaF₂:Mn dosimeters, except for a 5% loss in sensitivity after dark storage for 3 years at room temperature.

Suntharalingam et al. (7) have reported a gradual *loss* of sensitivity (as measured by light sum) in LiF (TLD-100) powder during predose storage at room temperature after annealing at 400°C for 1 hr and 80°C for 24 hr. They estimated this change at 5% in 12 weeks, and suggested that most of the decrease might occur during the first month after annealing. Using the same annealing procedure, the present LiF needles show a 7% loss in peak height during predose storage for 1 month, in rough agreement. Evidently the traps responsible for the No. 4 and No. 5 glow peaks are themselves not completely stable and are very gradually destroyed, or perhaps the Mg is precipitated out of the lattice as suggested by Claffy (6), during long storage periods. The presence of a large population of low-temperature traps, e.g., in rapidly cooled LiF, appears to supply new No. 4 and No. 5 traps more rapidly than they are destroyed, so the sensitivity of those glow peaks grows with time. In 400° + 80°-annealed LiF the population of low-temperature traps has been practically eliminated; hence the losses of No. 4 and No. 5 traps cannot be replaced, and the sensitivity decreases with time.

Change of Glow Curve on Storage After Irradiation

We have seen that there is a gradual change in glow-curve shape for LiF needles stored for various times between a prior readout and γ irradiation. An identical experiment was also done with the same batch b dosimeters, the only change being the shift of the ^{60}Co γ -ray exposure from the *end* of the storage period to the *beginning*, immediately after the prior readout operation. A second group of dosimeters, from batch a, was run for comparison to show the effect of having a smaller population of low-temperature traps. As before, the dosimeter readings after storage were compared to readings obtained after subsequent reexposure and prompt readout.

Results are shown in Fig. 4. Curve B reveals that the growth in the maximum glow-peak height is similar in the present case to that shown by Curve A, but somewhat smaller in magnitude. This similarity implies that a single phenomenon probably accounts for the growth in both cases; i.e., migration and aggregation of trapping centers. Note that this model requires that a substantial fraction of the filled traps retain their trapped charge carriers during postexposure migration and subsequent formation of the new, more complex, trap configurations. The fact that Curve B is lower than A suggests, however, that some loss of charge carriers does indeed occur during postexposure storage, while migration is taking place.

In comparing LiF batch b (Curve B) with batch a (Curve C) in Fig. 4, it is evident that C rises more rapidly at first, but B reaches higher values later. The larger population of low-temperature traps in batch b can explain the latter, but the reason for the early slower rise of Curve B is more obscure: In some of the batch b needles peak 3 was relatively more pronounced than shown in Curve A of Fig. 3 and was not clearly resolved from peaks 4 and 5. The decay of peak 3 during postdose storage thus *lowered* the maximum peak height slightly during the first few days for these individual needles. The effect of averaging these with the rest of the group, which showed a rise similar to Curve C, resulted in the apparent slow rise of Curve B. The more difficult question as to why individual dosimeters show such marked differences in their glow-curve shapes remains unanswered, at least for the present.

Curve C has been followed out to longer storage periods than shown in Fig. 4: It had dropped to 1.09 at 3 months, 1.05 at 1 year, and 1.00 at 3 years. This results from the combined effects of a decreasing population of the high-temperature traps 4 and 5 after the supply of low-temperature traps becomes exhausted, and leakage of charge carriers from high-temperature traps. According to the findings of Suntharalingam et al. (7) with LiF (TLD-100) powders annealed at 400°C for 1 hr and 80°C for 24 hr, approximately the same decrease (5%) in light sum occurred in 3 months whether the γ -ray exposure was given before or after the storage period. This implies that trap leakage has a relatively minor influence on the observed decrease of peaks 4 and 5 compared with the gradual elimination of the trapping centers themselves.

Since the sensitivity of $\text{CaF}_2:\text{Mn}$ has been found not to depend upon previous thermal history, the readings of dosimeters which have been cycled, exposed, then stored for various periods and read may be compared to readings obtained from dosimeters precycled at any time before exposure, exposed, and read promptly. The fading of the $\text{CaF}_2:\text{Mn}$ dosimeters with a γ -ray exposure of 1 R is shown in Fig. 4, Curve E. Fading is about 9% in one month, due entirely to leakage of charge carriers from traps. It is interesting to note that the fading in the first day is only about half of that observed for $\text{CaF}_2:\text{Mn}$ glass bulb dosimeters (8), probably because of the extended heating program used to read the needles (4 sec longer). Schulman et al. (8) have shown that the apparent peak-height fading in $\text{CaF}_2:\text{Mn}$ increases with the heating rate employed during readout.

Effects of Pre- and Postexposure Annealing at 100°C

We have found that for LiF dosimeters, preexposure or postexposure annealing at 100°C can produce about the same glow-curve changes as does storage at room temperature before or after exposure. The effect on the major peak height of the LiF \bar{b} dosimeters with pre- and postexposure annealing for various times at 100°C is shown in Fig. 5. As with room-temperature annealing of longer duration (Fig. 4), the growth of maximum peak height with annealing time at 100°C is strikingly similar whether the radiation exposure is given before or after the annealing period. Postexposure annealing again gives somewhat less growth than does preexposure annealing, presumably due to charge leakage. Gorbics and Attix (9) observed this growth when extruded LiF (TLD-700) samples were annealed for 15 min at 100°C after being γ irradiated, and attributed the effect to the escape of charge carriers from low-temperature traps with subsequent *retrapping* in the high-temperature ones. This explanation cannot, however, account for similar growth of the high-temperature peaks as a result of preexposure annealing, and therefore is probably incorrect. If retrapping is taking place, it must be a relatively minor influence. Mayhugh et al. (10) have arrived at the same conclusion based on similar observations.

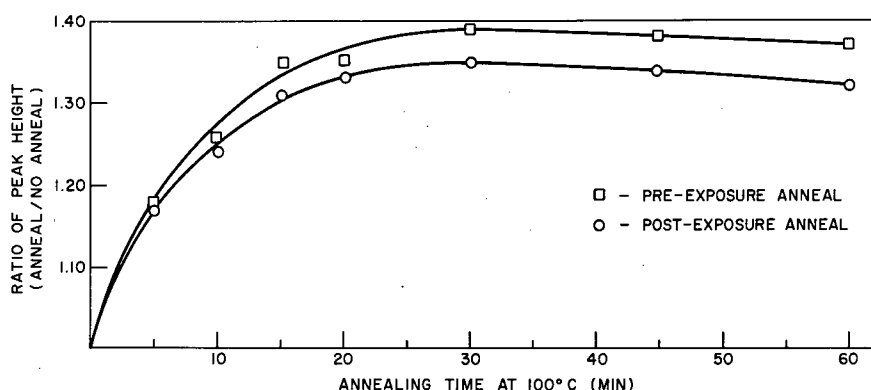


Fig. 5 - Growth of major peak height of LiF \bar{b} dosimeters expressed as the ratio, anneal/no-anneal, vs pre- and post-exposure annealing time at 100°C. Heating current was 4 A.

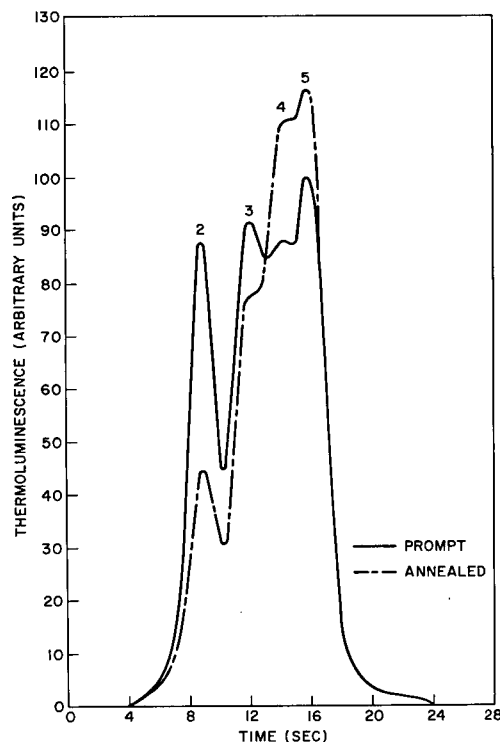
Figure 5 also corroborates the conclusion that, once the supply of low-temperature centers is exhausted and no more new high-temperature traps can be formed by aggregation, then the gradual decay of the latter traps causes the glow-peak maximum to decrease in amplitude.

The role of trap migration and the accumulation of simpler centers into more complex ones in LiF (TLD) has been discussed by Grant and Cameron (11), Claffy (6) in collaboration with J.H. Schulman, Christy et al. (12), Mayhugh et al. (10), and Jackson and Harris (13). Grant and Cameron identified the No. 2 glow peak with Mg^{2+} -positive-ion-vacancy dipoles, on the basis of dielectric-loss measurements. Jackson and Harris have concluded that the No. 3 glow-peak trap is also an integral part of this dipole center, and that peaks 2 and 3 are associated with two optical absorption-band components both located at 380 nm. This band was found to diminish during postexposure annealing at first rapidly, in correspondence with the reduction of the No. 2 TL glow-peak height, then more slowly, at the same rate peak 3 was reduced.

In a like manner, glow peaks 4 and 5 were identified with rapidly and slowly fading components of an optical absorption band at 310 nm. The center responsible for the No. 4 glow peak and the rapidly fading component of this band was suggested by Jackson and Harris to be a grouping of two or three Mg^{2+} -positive-ion-vacancy dipoles. However, a separate, unidentified trapping center was proposed to account for the slowly fading component of the 310-nm band, and the No. 5 glow peak. This complication was thought to be necessary to account for the fact that peak 5 was not observed by Jackson and Harris to grow during postexposure annealing at 40 to 90°C, although the 310-nm band showed growth corresponding to the rate of decay of glow peak 3. The traps responsible for glow-peak 2 were believed to lose their charge carriers rapidly during such annealing, and the associated dipole centers, with charge carriers still held in their No. 3 traps only, were pictured in this model as migrating together to form dimer or trimer centers related to the No. 4 glow peak only, but not No. 5.

In the present work, as also reported by Gorbics and Attix (9) and Mayhugh et al. (10), growth of glow peak 5 seems clearly present after suitable pre- or postexposure annealing. To verify this further we have slowed the heating rate slightly during readout to obtain clearer separation of the No. 4 and 5 peaks. Figure 6 shows the resulting glow curves for LiF b needles irradiated and promptly read out, and for ones annealed for 15 min at 100°C before irradiation and readout, with the heater current reduced to 3 A from the usual 4 A. It can be seen that both peaks 4 and 5 grow as Nos. 2 and 3 decay.

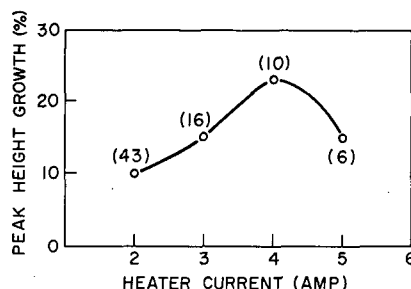
One possible explanation for the apparent absence of peak 5 growth during annealing in the experiment of Jackson and Harris may be that they employed such a slow heating rate (120°C/min) for the TL readout that the readout process itself served as a partial annealing treatment, a process which they postulated to explain the lack of a growth in peak 4 in their TL experiments. Thus there would be relatively less difference between the glow curves from "annealed" and "nonannealed" samples. Moreover the LiF batch employed by Jackson and Harris had less-pronounced low-temperature peaks, probably partially due to the slower cooling rate used. Hence, the growth to be expected in peaks 4 and 5 due to annealing would have been correspondingly less.



These effects are illustrated in Fig. 7, which shows, for various heating currents, the percentage of growth in the major glow-peak height for LiF needles given a 15-min postexposure anneal at 100°C, relative to readings without annealing. As usual, all dosimeters had been given a normal 4-A readout cycle before exposure to γ rays. The dosimeters chosen for this test showed somewhat less low-temperature peak height than the typical needles of batch b; thus the maximum peak-height growth is only 23% at 4 A, as compared with a corresponding value of 30%

Fig. 6 - Glow curves, thermoluminescence vs time, for LiF b dosimeters read promptly after exposure, and for those given an anneal of 100°C for 15 min prior to exposure and readout. Heater current is 3 A.

Fig. 7 - Curve showing peak-height growth of LiF dosimeters given a postexposure anneal of 100°C for 15 min relative to unannealed dosimeters vs heater current. Numbers in parentheses are the times (in seconds) from commencement of heating cycle to readout of major peak height.



in Fig. 5. At lower currents the growth is observed to be less than at 4 A, as predicted. However, anomalous behavior occurs at a 5-A heating current, resulting in a rapid reduction in peak growth. We will not attempt to explain this in detail, except to point out that all resolution between glow peaks is lost at this very rapid heating rate ($\sim 2000^\circ\text{C}/\text{min}$); thus, even peak 2 could be contributing to the maximum peak height in the unannealed dosimeters. Moreover, the effect of thermal quenching (8) should decrease peaks 4 and 5 more than peaks 2 and 3, and the annealed dosimeters should suffer the more pronounced peak-height reduction on this account since only peaks 4 and 5 are present.

With respect to the specific model of LiF thermoluminescence described by Jackson and Harris (13), our findings tend to support it in principle, except that the growth we have observed in glow peak 5, as well as 4, suggests that the trapping centers responsible for both of these peaks are being formed by the migration and aggregation of simpler centers during annealing at moderate temperatures. Peaks 4 and 5 are not necessarily related to identical centers, however, as pointed out by Jackson and Harris.

SUMMARY OF STORAGE DATA

It is interesting at this point to bring together the room-temperature storage data expressed in a somewhat different form, along with other data to show how prereadout annealing at 100°C for 15 min can be used to reduce the possible dosimetry errors which could occur because of the change in glow-curve shape and peak height during storage since the last readout. In Table 1 we show the storage, annealing, γ -exposure, and readout histories of various groups of dosimeters (10 each) from Day 0, when all were cycled through the reader, until they were finally read out. The peak-height readout results are given in parentheses, normalized to the average reading of the same dosimeters which have been cycled and exposed on Day 0 and read 32 days later with no pre- or postexposure annealing. Note that prereadout annealing at 100°C for 15 min (Runs 12 to 18) results in a readout within $\pm 3\%$ of that obtained in the reference case (Run 6), regardless of when the exposure or reading occurred. This procedure therefore offers a satisfactory alternative to the usual method of preexposure annealing (1 hr at 400°C and 24 hr at 80°C) in routine dosimetry applications. It should be remembered, however, that total exposures exceeding about 10^3 R will result in an increase in phosphor sensitivity that a simple readout cycle will not remove. Annealing at 400°C for 1 hr, followed by a readout cycle, is required in that case to restore the LiF to its original sensitivity.

Several additional performance tests of these dosimeters were undertaken to determine some of their other characteristics. These are discussed in the following sections.

Table 1
Summary of LiF b Needle Readings vs Storage Time in Darkness at 22°C,
for Various Treatment Procedures

Run Number	Day 0	Day 1	Day 4	Day 7	Day 15	Day 32
1	CDR(0.75)					
2	CD	R(0.78)				
3	CD		R(0.84)			
4	CD			R(0.88)		
5	CD				R(0.94)	
6	CD					R(1.00)
7	C	DR(0.78)				
8	C		DR(0.85)			
9	C			DR(0.92)		
10	C				DR(0.98)	
11	C					DR(1.03)
12	CDAR(0.97)					
13	CD	AR(0.97)				
14	CD			AR(0.98)		
15	CD					AR(0.99)
16	C	DAR(1.01)				
17	C			DAR(1.02)		
18	C					DAR(1.03)

Explanation of symbols:

C — cycled through the TLD reader

D — dosed by exposure to 12.5 R ^{60}Co γ -radiation

A — annealed for 15 min at 100°C

R — read in the TLD reader

Example: In Run No. 14 the dosimeters were cycled through the reader and dosed on Day 0, then stored until the 7th day when they were annealed for 15 min at 100°C and then read out. The observed peak height was 2% less than in Run 6, to which the other runs are normalized.

Signal Erasure

Five dosimeters each of $\text{CaF}_2:\text{Mn}$ and ^7LiF were given ^{60}Co γ -ray exposures of 10 R in 10 min and read in the normal manner. After being cooled, these dosimeters were reread to determine what fraction of the TL signal remained. For both groups of dosimeters the remaining signal was less than 10 mR, indicating that no significant signal remained after readout for any exposure likely to occur in personnel monitoring operations.

Effect of Extended 100°C Annealing

During the 100°C annealing studies it was found that extended annealing periods caused slight reductions in the γ -ray sensitivity of the LiF dosimeters, amounting to nearly 5% after 5 hr. The dosimeters were found to return to their original sensitivity after being heated to 400°C for 1 hr, cooled to room temperature in 1 to 5 min, and then cycled once through the reader. Five cumulative hours of 100°C annealing with readout cycling every 15 min did not alter the sensitivity within experimental error (~2%).

Effect of Repeated Exposures and Readouts

The second test was conducted to determine if repeated exposures given promptly would change the response of the dosimeters. For this test, five $\text{CaF}_2:\text{Mn}$ dosimeters and five ^7LiF dosimeters were given 50 consecutive exposures varying from 20 mR to 1 R of ^{60}Co γ radiation, with readout after each exposure. At the beginning of the experiment and after every tenth exposure each group of dosimeters was given a "test" γ -ray exposure of 12 R. Both the peak height and area were the same for each test exposure within $\pm 2\%$ for both groups. In addition, no change in the glow-curve shape was noted for any of the test exposures.

Effect of 18 Months in Routine Application

A third test was attempted to determine if continued use in personnel monitoring would change the response of the dosimeters if no annealing procedures were used. Five each of the $\text{CaF}_2:\text{Mn}$ and ^7LiF needles were studied after they had been used in routine personnel monitoring operations for 18 months. They had been read monthly during that period, and had indicated exposures ranging from 20 mR to 800 mR per month. After being routinely read for the 18th month, each dosimeter was given a prompt "test" exposure of 12.5 R of ^{60}Co γ radiation and read. All the dosimeters were then annealed for 1 hr at 400°C, cooled, and then cycled through the reader. It was assumed that this annealing procedure returned the LiF dosimeters to their original response characteristics, as is demonstrably the case after damage due to extended 100°C annealing, as mentioned earlier. All the dosimeters were then given another "test" exposure of 12.5 R and read. The latter "test" exposure readings, which presumably were the same as those at the beginning of the 18-month period, were not significantly different from the ones given at the end of the 18-month period before the 400°C, 1 hr annealing treatment. The peak height and area of the $\text{CaF}_2:\text{Mn}$ dosimeters were changed by $< 3\%$. The peak height of the ^7LiF dosimeter test exposure given after the 400°C annealing procedure was about 2% less than that before, while the area increased by ~6%. This increase in area was caused by an increase in peak 6 at the high-temperature end of the glow curve. No other changes in glow-curve shape were observed.

This same experiment was repeated using dosimeters of both kinds which had been stored at room temperature for 3 years since the last readout, with slightly better

results: no change in peak height or area, $\pm 2\%$. Apparently, cycling the dosimeters through the reader returns them consistently to the same response characteristics after almost any period of annealing at room temperature and even after many readings. Eastes (14) has reported similar results using ^7LiF needle dosimeters which were given calibration exposures up to 10 R weekly for 90 consecutive weeks. The dosimeters, which were given no formal annealing procedures, showed no change in response for the duration of the experiment.

Effect of 15-Min, 100°C Postexposure Annealing

A group of ten ^6LiF dosimeters were given 20 consecutive exposures of 12.5 R of ^{60}Co γ radiation. After every second exposure the dosimeters were annealed at 100°C for 15 min before readout. The individual readings of the ten exposures followed by annealing varied from the average of the same readings with a standard deviation of only $\pm 1.1\%$. The exposures without postexposure annealing were almost as consistent, showing a standard deviation of only $\pm 1.3\%$ from the average. We then repeated the experiment with all exposures being followed by a 15-min postexposure anneal. These readouts were even more consistent, the standard deviation from the average being only $\pm 0.8\%$. No changes in glow-curve shape were observed during these experiments.

Precision

Ten dosimeters each of ^7LiF and $\text{CaF}_2:\text{Mn}$ were given ten consecutive ^{60}Co γ -ray exposures of 20 mR and read using the Honeywell Electronik 19 recorder to determine their reproducibility from exposure to exposure. The standard deviations of the individual dosimeters from the average of each set of readings averaged $\pm 7.0\%$ for the ^7LiF dosimeters and $\pm 7.9\%$ for the $\text{CaF}_2:\text{Mn}$ dosimeters. Ten exposures at 90 mR resulted in average standard deviations of $\pm 3.4\%$ for the ^7LiF and $\pm 2.8\%$ for the $\text{CaF}_2:\text{Mn}$ dosimeters when the dosimeters were read using the Electronik 19 recorder. When read on the built-in recorder the standard deviations at 90 mR averaged $\pm 5.9\%$ for ^7LiF and $\pm 5.8\%$ for $\text{CaF}_2:\text{Mn}$. At higher exposures reproducibilities of $\pm 2\%$ to $\pm 5\%$ were achieved, depending on where the readouts occurred on the recorder scales.

Another important aspect of the precision of the system is the variation in response per roentgen from dosimeter to dosimeter in a group, since this determines whether individual correction factors must be used in evaluating results obtained from the dosimeters. To test this, 50 $\text{CaF}_2:\text{Mn}$ dosimeters were given a ^{60}Co exposure of 140 mR and read using the built-in recorder. The standard deviation of the individual dosimeter readings from the average reading was $\pm 7.1\%$. Two groups of LiF dosimeters (100 each, ^6LiF and ^7LiF) were exposed to 400 mR ^{60}Co γ radiation and read using the built-in recorder. They indicated even less variation from dosimeter to dosimeter, the standard deviation of the ^6LiF and ^7LiF dosimeters being only $\pm 4.8\%$ and $\pm 3.2\%$, respectively, from the averages. Two groups of 50 each of ^6LiF and ^7LiF obtained later had standard deviations of ± 7.2 and $\pm 4.8\%$, respectively, from the averages. Obviously, no correction factors for individual dosimeters would normally be required for ordinary personnel-monitoring applications using these dosimeters.

Built-In Background

Previous studies of larger TL-32 type EG&G glass-bulb TLD's (15) have shown that built-in background due to radioactive contamination in the TL phosphor and/or the glass may amount to 1 mR per day. This results in an inconvenience for processing personnel-monitoring data, since the background must be subtracted from the total indicated

exposure. To determine the built-in background of the glass-needle dosimeters, ten dosimeters each of $\text{CaF}_2:\text{Mn}$, ^6LiF , and ^7LiF were stored in the dark at room temperature ($\sim 22^\circ\text{C}$) for 7 months in a low-background office area and then read. The readings were corrected for fading (using the data for 1 month) and for differences in individual response of the dosimeters. The $\text{CaF}_2:\text{Mn}$ dosimeters indicated an average background of 4.6 mR/mo; the ^7LiF dosimeters, 4.4 mR/mo; and the ^6LiF dosimeters, 5.0 mR/mo. Since external background is believed to be about 4 mR/mo for this office area, it is concluded that there is no significant background due to radioactive contamination in the TL phosphor or the encapsulating glass tube.

Response to Beta and X Radiation

The response of the TL needles to beta radiation was determined by exposing ten $\text{CaF}_2:\text{Mn}$ and ten ^7LiF needles bare and in the finger ring to the betas from natural uranium disks. Two-hour exposures were made in contact with the uranium disks. The contact dose rate in tissue from the disks was assumed to be 228 mRad per hour (16). The average dose response of the bare $\text{CaF}_2:\text{Mn}$ dosimeters to these betas relative to the ^{60}Co γ -ray exposure response was 0.72 R/rad, with a standard deviation of $\pm 3.8\%$. That is, one tissue rad of β rays gives the same response as does 0.72 roentgens of ^{60}Co γ rays. The response of the bare ^7LiF needles was 0.68 R/rad, also with a standard deviation of $\pm 3.8\%$. When exposed in the finger rings the response of the $\text{CaF}_2:\text{Mn}$ needles is reduced to $0.25 \text{ R/rad} \pm 22\% \text{ S.D.}$, and the response of the ^7LiF needles is reduced to $0.23 \text{ R/rad} \pm 21\% \text{ S.D.}$

As an aid to determining the effective energy of the radiation encountered in personnel monitoring, the response ratio CaF_2/LiF of the needles was determined using four energies — 38, 70, 117, and 169 keV — of heavily filtered x rays. For this test five finger rings, containing one each of ^7LiF and $\text{CaF}_2:\text{Mn}$ dosimeters, were exposed to approximately 1 R of x radiation at each of the four energies. All dosimeters were read and the readings normalized to the ^{60}Co response of each individual dosimeter. The CaF_2/LiF response ratio at the four energies were: 8.90 at 38 keV, 5.50 at 70 keV, 2.10 at 117 keV, and 1.26 at 169 keV. These data, along with the calculated response ratio, are shown in Fig. 8. The calculated response was computed from the mass energy-absorption coefficients (μ_{en}/ρ) of the phosphors, and normalized to the measured data at 1.25 MeV.

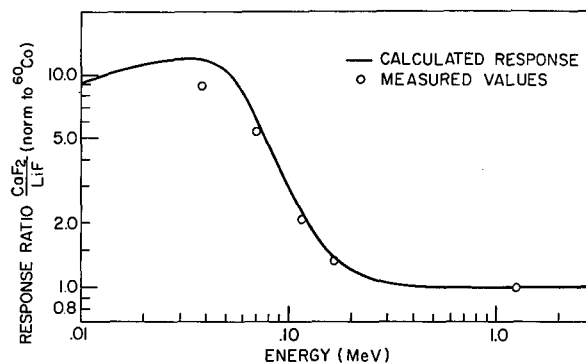


Fig. 8 - Response ratio, $\text{CaF}_2:\text{Mn}/\text{LiF}$, vs γ -ray energy, normalized to ^{60}Co γ radiation

CALIBRATION PROCEDURES

We calibrate our reader to minimize the effects of changes in peak height and stored signal fading discussed previously. For the LiF dosimeters we give a group of standard dosimeters (dosimeters whose γ -ray response is the same as the average of the group) a known ^{60}Co γ -ray exposure followed by a 15-min anneal at 100°C before readout. The correction factor for the average of the group is determined from these readings. From Table 1 we see that the exposures determined from readings of dosimeters which have been worn for one month and read with no postexposure annealing will never be underestimated using this procedure. The maximum overestimation would be 6%. If for any reason a dosimeter is read before the end of a month's wearing period, we also give it an anneal of 100°C for 15 min before readout to eliminate errors due to glow curve change or stored signal fading, as shown in Table 1. This procedure is also employed for dosimeters which are used for other applications such as x-ray surveys, where the time since the last readout may be only a few hours or days.

For $\text{CaF}_2:\text{Mn}$ dosimeters we also use standard dosimeters which are given a known ^{60}Co γ -ray exposure to determine the correction factor for the group. To account for stored signal fading in dosimeters worn for one month, the readings of these promptly read dosimeters are reduced 10% before determination of the correction factor to be used. For dosimeters read on a monthly basis this procedure results in an overestimation of that part of the exposure received during the last five days before readout. If one assumes that exposures occur uniformly throughout the month, the overestimation of exposures will only be a few percent and of course will be 10% only if all the exposures were received on the last day before readout.

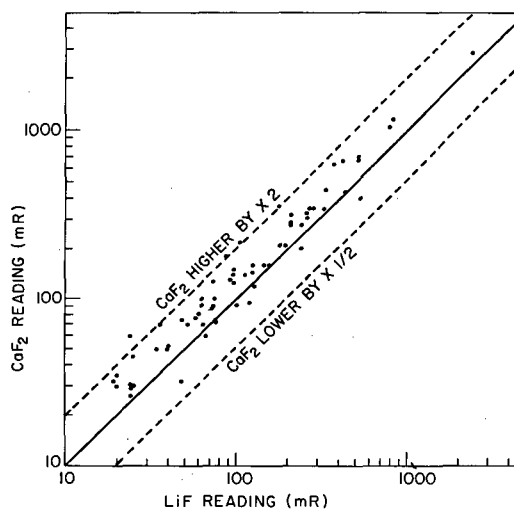
RESULTS OF ROUTINE PERSONNEL-MONITORING TESTS

Use of the TL needles in routine personnel monitoring in the plastic finger rings began in the Spring of 1967 and has continued uninterruptedly since that time. The performance of the TL needles has been entirely satisfactory. Occasional breakage of one, but never both of the needles, has occurred. No difficulty in reading the dosimeters has been encountered. In normal use rings are collected, the needles read and returned to the same ring, and the ring returned to the same person. In evaluating the results from the rings no correction for individual response of the dosimeters is used, only the correction factor for the average of the group. No attempt has been made to compare the TL system to film, since we have found the film to be completely unsatisfactory for this application, primarily due to light leakage. However, the agreement between the CaF_2 reading and the LiF reading for each ring has generally been good. In Fig. 9 we have plotted CaF_2 reading vs LiF reading for 65 rings processed over a three-month period in 1967. The CaF_2 readings average higher than the LiF readings, the average ratio CaF_2/LiF being 1.35, indicating some exposure to low-energy γ radiation. The standard deviation of the ratios is $\pm 25\%$. Since each finger ring was not necessarily exposed to the same energy radiation, this standard deviation is not an indication of the accuracy of either the CaF_2 or LiF dosimeters, but it does set an upper limit on the magnitude of the errors inherent in the dosimetry system.

CONCLUSIONS

Our laboratory and field tests of glass capillary "needle" TLD's have shown them to be very useful for personnel monitoring and other routine Health Physics applications. We have found that they can be used without any formal annealing procedures, provided they are read on a regular schedule. We found that the peak height readout of the LiF dosimeters varied considerably with time since the last readout, but that significant

Fig. 9 - $\text{CaF}_2:\text{Mn}$ dosimeter reading vs LiF dosimeter reading for 65 finger rings worn at the NRL Linac



errors could be eliminated by giving the dosimeters an anneal of 100°C for 15 min before readout. We have successfully used these dosimeters in finger-ring applications since 1967 and routinely use them for other Health Physics applications. Because of their ease of handling, with no necessity for complicated annealing, and the rapidity with which results can be obtained after use, these dosimeters have gained wide acceptance by the field health physicists at NRL for many monitoring applications where film was formerly used.

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13. ABSTRACT			
<p>Miniature glass-encapsulated needle-shaped thermoluminescent (TL) dosimeters, 1.4 mm in outer diameter and 12 mm in length, have been investigated for routine application in finger rings to monitor the dose received by the hands, e.g., in certain Linac repair operations. Both $\text{CaF}_2:\text{Mn}$ and LiF-filled dosimeters were studied, and special attention was given to the simplification or elimination of the cumbersome annealing procedures normally used with LiF dosimeters.</p> <p>It was found that the heights of the LiF TL glow peaks in the vicinity of 200°C, usually employed for dosimetry, display a marked growth as a function of storage time occurring either before or after a given γ-ray exposure. This is probably caused by the migration and accumulation of simple, low-temperature trapping centers into more complex, higher-temperature centers, as suggested by some other investigators. The magnitude of the effect depends on the population of low-temperature traps present, and on the readout heating rate employed. It was found possible to reduce the dosimetry error caused by this sensitivity increase to negligible levels ($\pm 3\%$) through the use of a routine, prereadout annealing treatment of 15 min at 100°C. No additional annealing procedure was found to be necessary for LiF, so long as the γ-ray exposures were not large enough ($\sim 10^3$ R) to cause radiation-induced sensitization of the phosphor. $\text{CaF}_2:\text{Mn}$ showed no significant change in sensitivity either upon storage or radiation exposure.</p>			

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	ROLE	WT	ROLE	WT	ROLE	WT
Dosimeters, luminescent Dosimetry Dosimetry, radiation						